Irradiation Creep and Swelling of Russian Ferritic-Martensitic Steels Irradiated to Very High Exposures in the BN-350 Fast Reactor at 305ºC -335ºC

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Abstract: Russian ferritic/martensitic (F/M) steels EP-450, EP-852 and EP-823 were irradiated in the BN-350 fast reactor in the form of gas-pressurized creep tubes. The first steel is used in Russia for hexagonal wrappers in fast reactors. The other steels were developed for compatibility with Pb-Bi coolants and serve to enhance our understanding of the general behavior of this class of steels.

In an earlier paper we published data on irradiation creep of EP-450 and EP-823 at temperatures between 390 and 520°C, with dpa levels ranging from 20 to 60 dpa. In the current paper new data on the irradiation creep and swelling of EP-450 and EP-852 at temperatures between 305 and 335°C and doses ranging from 61 to 89 dpa are presented. Where comparisons are possible, it appears that these steels exhibit behavior that is very consistent with that of Western steels. Swelling is relatively low at high neutron exposure and confined to temperatures <420°C, but may be camouflaged somewhat by precipitation-related densification. These irradiation creep studies confirm that the creep compliance of F/M steels is about one-half that of austenitic steels.

Keywords: ferritic/martensitic steels, irradiation creep, swelling.

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Introduction

Ferritic/martensitic (F/M) steels are widely used as structural materials in various types of reactor facilities. The main advantages of F/M steels are their high resistance to void swelling, low irradiation creep rates and a relatively low radioactivation after neutron irradiation. At the same time, the well-known disadvantages of these steels are their low long-term creep strength at high temperatures and their inclination to low-temperature irradiation embrittlement.

Earlier measurements of irradiation creep and short-term mechanical properties were performed as part of the current effort for two Russian F/M steels designated EP-450 (12Cr-1.3Mo-2V-Nb-B) and EP-823 (11Cr-1Mo-1Si-Nb, V, W) [1]. They were irradiated in the BN-350 fast reactor to doses of 20-60 dpa and have demonstrated that at irradiation temperatures below ~500°C the irradiation creep rate in the steels is rather low and consistent with measurements made on various Western F/M steels over the same temperature range.

Some results of this earlier study are shown in Figure 1 and demonstrate that not all strains measured in creep tests arise from irradiation creep alone. Note that at the irradiation temperature of 520°C an apparent increase of irradiation creep modulus is observed due to the onset of thermal creep and concurrent loss of strength. Note also that the creep modulus appears to increase as swelling begins at lower temperatures. The data are also interpreted to show negative precipitation-related strains that lower the apparent creep modulus when the irradiation creep component is relatively small.

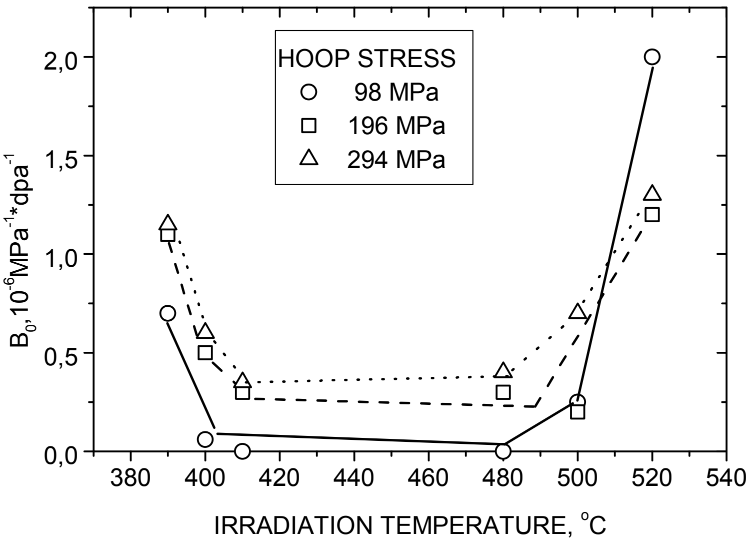

FIG. 1 - *Average creep coefficients derived for EP-450 in the range 390-520°C for three levels of hoop stress [1]. The data are interpreted to show the combined influence of swelling at low temperature, thermally assisted creep at higher temperature and precipitate-related densification over the entire temperature range. Similar behavior was observed in EP-823 at 390 and 480°C.*

However, the most sought-after data for such steels are those at relatively low temperatures. Acquisition of such data require that the irradiation be performed in a reactor with a relatively low inlet coolant temperature, such as found in the BN-350 fast reactor in Kazakhstan but not available in Western reactors. In the present paper results are presented of further investigation of irradiation creep and swelling in EP-450 and EP-852 ferritic-martensitic steels. These steels were irradiated as pressurized creep tube cladding in the reactor BN-350 at temperatures in the range of 305-335°C to a maximum dose of 89 dpa.

Experimental Details

The measured chemical composition and final heat treatment of the creep tubes made of the EP-450 and EP-852 F/M steels are shown in Table 1. Creep tubes of 6.9 mm external diameter and 0.4 mm wall thickness (Figure 2) were used. To produce hoop stresses in the range 0 - 250 MPa at irradiation temperatures of 305-335°C, the tubes were filled with argon of 99.998% purity through a needle valve located in the large blank end flange.

TABLE 1 - *The chemical composition of the EP-450 and EP-852 F/M steels*

Steel	Content, wt.%										
	C	Si	Mn	S	P	Cr	Ni	Mo	Nb	V	B
EP-450	0.14	0.20	0.31	0.009	0.017	12.95	0.20	1.54	0.47	0.22	0.004
	Solution treated 1050°C, 1 s + aged 850°C, 5 s.										
EP-852	0.13	1.91	0.31	0.009	0.017	13.15	0.27	1.69	-	-	-
	Solution treated 1050°C, 1 h + aged 720°C, 1 h.										

To reach high damage doses, the creep tubes were irradiated in the BN-350 reactor in special experimental subassemblies having extractable containers. These subassemblies are similar to regular driver subassemblies of the BN-350 reactor, but with 31 central pins replaced by an extractable cylindrical container of 32 mm in diameter. In each container, perforated cylindrical canisters were placed at different heights (Figure 3), with each of the canisters containing seven gas-filled tubes (one tube with zero gas pressure and six gas pressurized tubes, two tubes for each of three nominal hoop stress levels). The canisters were 97 mm in length, 26 mm in outer diameter and with 0.3 mm wall thickness. The required irradiation temperature for the creep tubes was ensured due to heating the container by surrounding pins. The calculated irradiation temperatures and doses for each container depend on the location of the canister with respect to the reactor core midplane. After irradiation in four consecutive reactor runs the containers were extracted from the spent subassemblies and inserted in fresh subassemblies, which then were placed at the same positions in the core. Afterwards, the irradiation of the samples was continued for an additional four runs. The final calculated doses and irradiation temperatures for the steels investigated are shown in Tables 2 and 3.

The surfaces of the irradiated creep tubes were cleaned in 50 % ethanol solution, and then the tube diameters were measured by a micrometer with an accuracy of 0.01 mm. For each tube the measurements were made at three cross sections: in the middle and at 15 mm apart from both tube ends, for two tube orientations that differ by rotation around the tube axis by 90°.

There were some gradients in dose and temperature along the tubes. For steels EP-852 (305°C/69 dpa), EP-450 (320°C/81 dpa) and EP-450 (335°C/89 dpa) the calculated gradient of dose was equal to 0.1 dpa/mm and the calculated gradient of temperature was 0.15°C/mm. For the distance of 50 mm between two cross sections at which tube diameters were measured, the difference of doses equals 5 dpa, and the difference of temperature was equal to 7.5°C. For EP-852 (310°C/61 dpa) and EP-450 (310°C/61 dpa) steels the calculated gradients of damage dose and of temperature were equal to 0.12 dpa/mm and 0.2°C/mm, respectively, so the doses and the temperatures vary by 6 dpa and 10°C, respectively.

For testing to ensure that the tube did not release its fill gas and for determining the actual hoop stresses, the tubes were punctured at room temperature in a remote installation and the volume of the gas was measured. Hoop stresses at the end of irradiation were calculated from the following equation.

$$\sigma_\theta = P(T_0 / T)(V / V_0) d_{\text{int}} / 2t \qquad (1)$$

where T is the temperature of the gas released from a punctured creep tube, P is the pressure of the released gas at temperature "T", V is the volume of the released gas at temperature "T", T_o is the irradiation temperature, V_0 is the internal volume of the creep tube at temperature "T_o", $d_{int.}$ is the internal tube diameter, and t is the tube wall thickness. The total irradiation creep strain ε^{ic} was determined as the difference between the total diametral strain and strain due to swelling (the diametral strain of the stress-free tube).

The irradiation creep modulus was calculated from the following equation, where the stress is the hoop stress.

$$B_0 = \varepsilon^{ic} / 0.75\sigma_0 \times dpa \qquad (2)$$

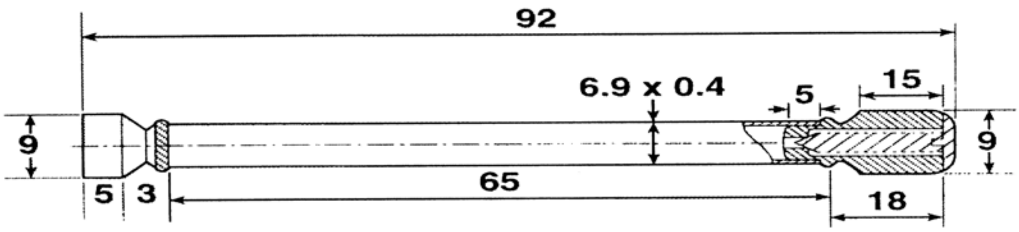

FIG. 2 - Irradiation *creep tube (all sizes in mm.)*.

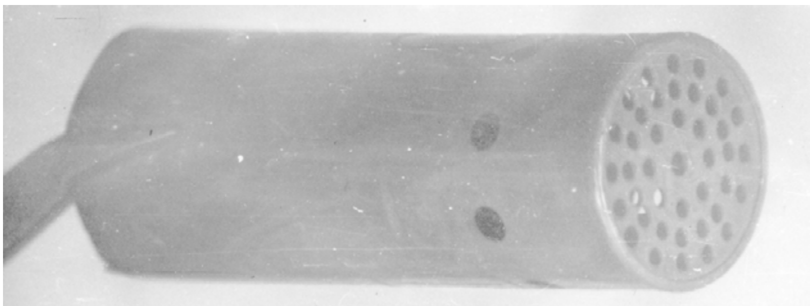

FIG. 3 - View *of the canister for containing the creep tubes*.

Results

Upon removal of the tubes from the various canisters, a visual inspection of the creep tubes did not reveal any surface defects. All irradiated tubes retained their initial shape and appearance. As a result of mechanical loading, however, five creep tubes, namely, one tube of the EP-450 steel and four tubes of EP-852 steel were destroyed while puncturing for gas release, with the large head sections breaking off. Two such brittle creep tubes are shown in Figure 4.

The gas volume measurement data expressed as applied hoop stresses are shown in Tables 2 and 3. As follows from these data, some creep tubes were found to have lost all or part of their fill gas before puncturing. This required some judgment to determine what stress level to use in the data analysis. Based on the measured strains some tubes obviously lost their pressure early and were treated as stress-free. Others were treated as having maintained their design nominal pressure until the final stages of their extraction from the reactor.

a)

b)

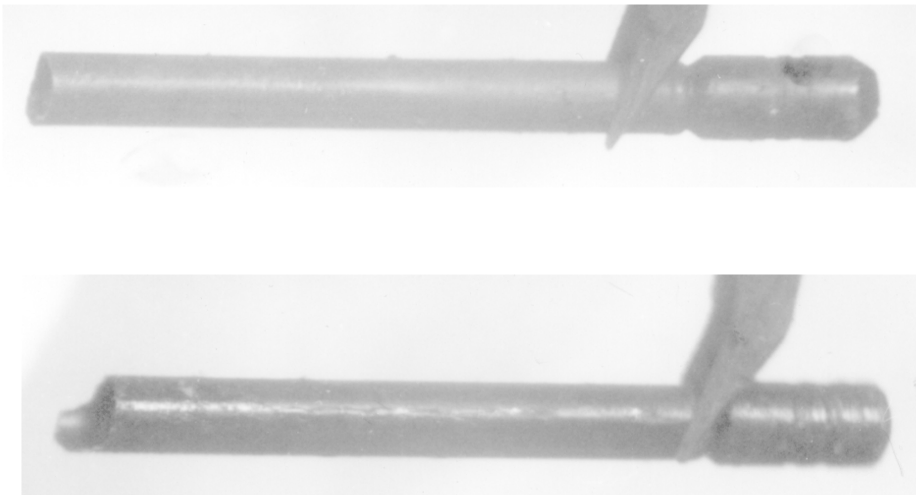

FIG. 4 - View *of failed creep tubes of the EP-450 steel (a) and EP-852 steel (b), both irradiated at the temperature of 310°C to 61 dpa.*

The results of measuring total creep tube diametral strains, irradiation creep strains and calculated magnitudes of the irradiation creep modulus in EP-450 and EP-852 steels are also shown in Tables 2 and 3. No systematic trend in the variation of tube diameter along the tube length was observed for EP-852 (310°C/61 dpa) and EP-450 (310°C/61dpa) steels. For EP-852 (305°C/69dpa) and EP-450 (320°C/81dpa, 335°C/89dpa) steels the tube diameter near the large plug (the top of the tube) was found to be slightly larger than near the small plug end.

From data on the diameter of stress-free tubes it follows, that the diameter change due to swelling is negligible in tubes of both steels. For stress-free creep tubes made of the EP-450 steel irradiated at 320°C and 335°C the diameter did not change under irradiation. For the tube of this steel irradiated at 310°C the diametral strain is equal to 0.1 %, but that does not exceed measurement errors. The same magnitude of the diametral strain (0.1 %) was measured for EP-852 steel irradiated at 310°C. Comparison with the results of the previous study [1], swelling of these steels appears to peak in the vicinity of 400ºC.

Thus, as a first approximation it is reasonable to assume that the diametral strain of creep tubes investigated near 300ºC is equal to the irradiation creep strain. The final conclusion concerning the balance of swelling, creep and densification strains will be made after TEM examination of the irradiated creep tubes.

One can see from the data shown in Table 2 that for the EP-450 steel the maximum irradiation creep strain is equal to 1.1% at the irradiation temperature of 310°C and at the dose of 61 dpa. At the hoop stress of 200 MPa the increase of both the irradiation temperature and dose does not result in an increase of the irradiation creep strain. The plot of the irradiation creep strain versus hoop stress is linear in EP-450 steel at all three irradiation temperatures investigated with some possible indication that precipitate-related strains may be included (see Figure 5).

The maximum irradiation creep strain of the EP-852 steel does not exceed 0.45 % and appears to be linear with stress. However, a full comparison between the two steels is somewhat impeded due to gas leakage or failure of several creep tubes with EP-852 steel cladding irradiated at 310°C to 61 dpa. At the irradiation temperature of 305 °C and the dose of 69 dpa the irradiation creep strain of EP-852 steel is relatively small (see also Figure 5).

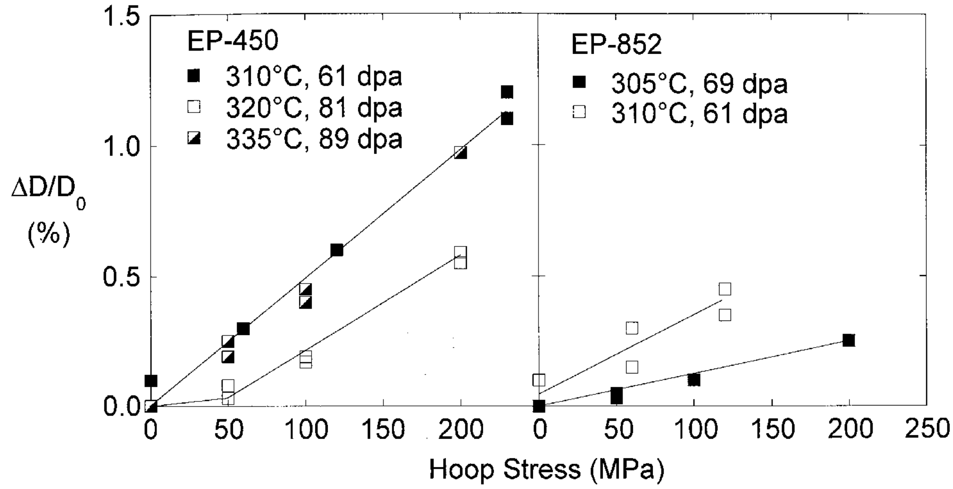

FIG. 5 - Irradiation *creep strain versus nominal hoop stress for creep tubes made of EP-450 and EP-852 F/M steels irradiated in BN-350 fast reactor*.

Discussion

At present, experimental data on irradiation creep characteristics of F/M steels are relatively scarce. In references 2 and 3 cladding creep tubes from the HT-9 F/M steel irradiated in the FFTF up to the high dose of 208 dpa have been investigated. French ferritic-martensitic steels EM-10 and EM-12 were irradiated to 77 dpa in PHENIX as creep tubes from typical cladding [4]. In the experiments mentioned above the minimum irradiation temperature was equal to 400°C. Lower irradiation temperatures are not available in the FFTF and PHENIX reactors because of their high inlet coolant temperatures. The inlet temperature of sodium in the BN-350 reactor is lower (~280°C) and allows irradiation of samples at much lower temperatures in comparison with those of other fast reactors.

From the data of the present work (see Table 2) one can see that for EP-450 steel the maximum value of the irradiation creep modulus B equals 0.74×10^{-6} $(\mathrm{MPa}\times\mathrm{dpa})^{-1}$ at irradiation temperatures in the 310-335°C range. Higher B values of $(1.6–1.7)\times10^{-6}$ $(\mathrm{MPa}\times\mathrm{dpa})^{-1}$ for several creep tubes should be ignored because the measured gas pressure in these tubes was found to be much lower than the initial pressure. If we assume that these tubes have lost part of the gas inventory after termination of the irradiation and that during irradiation the gas pressure in the tubes was equal to the nominal pressure, the values of *B* for these tubes will be equal to (0,91-1,0)$\times 10^{-6}$ (MPa$\times$dpa)$^{-1}$. For the EP-852 steel the nominal and measured hoop stresses differ insignificantly, and the calculated value of *B* does not exceed 0.75$\times 10^{-6}$ (MPa$\times$dpa)$^{-1}$.

By comparing these values of irradiation creep moduli with the results of previous investigations [2-4] one can conclude that lowering the irradiation temperature from 400°C to 305-335°C does not lead to a significant change of irradiation creep rate in F/M steels.

The design of experimental subassemblies in the present experiment did not provide an opportunity to perform intermediate measurements of creep tube diameters during the course of irradiation. Therefore, it was not possible to determine the dependence of irradiation creep strain on dose for the EP-450 and EP-852 steels. As for the irradiation creep strain dependence on applied stress, this dependence is approximately linear in the EP-450 and EP-852 steels at all irradiation temperatures investigated (Figure 5). It was anticipated that there might be a deviation from linearity at higher stress levels, as sometimes observed at higher irradiation temperatures in other F/M steels.

For example, in HT-9 investigated in reference 2 the transition from linear behavior to a power law dependence with the exponent n>1 has been observed at ~400ºC for hoop stresses in the range of 150-200 MPa. High values of n are typical following the onset of thermal creep, where the stress dependence of thermal creep rate can be described by a power law with exponent of 4 and higher [5]. Thus, most likely, an increase of n from unity beginning from some critical hoop stress provides evidence of a change of irradiation creep mechanism [6]. In the present case, however, the thermal creep is negligible at irradiation temperatures of 305-335°C, as this fact was confirmed earlier in special thermal creep tests conducted on these tubes.

Conclusions

Measurements of stress-free swelling and irradiation creep strains in EP-450 and EP-852 F/M creep tubes irradiated in BN-350 at temperatures of 305-335°C to doses of 61-89 dpa allow us to draw the following conclusions.

1. Neutron irradiation at conditions mentioned above results in a strong degradation of ductility of the steels. While puncturing to release gas one of the gas creep tubes of EP-450 steel and four tubes of EP-852 irradiated at 310°C to 61 dpa failed in a very brittle mode.
2. Swelling and possibly precipitate-related densification of the EP-450 and EP-852 steels determined by measuring the stress-free creep tube diameter are very small in the vicinity of 300°C, falling within the measurement accuracy. Comparing with the results of the previous study, swelling of these steels appears to peak in the vicinity of 400ºC.
3. The irradiation creep strain dependence on hoop stress of these steels is essentially linear at the irradiation conditions investigated.
4. At irradiation temperatures of 305-335°C and doses of 61-89 dpa the magnitude of irradiation creep modulus *B* for the EP-450 and EP-852 steels does not exceed 1.0$\times 10^{-6}$ (MPa$\times$dpa)$^{-1}$, confirming the relative resistance of F/M steels to irradiation creep at these low irradiation temperatures.

TABLE 2 - *Irradiation creep characteristics of EP-450 F/M steel*

310°C/61 dpa				320°C/81 dpa				335°C/89 dpa			
σ_θ, МПа measured (nominal)	**Δd/d, %**	**ε^{ic}., %**	**B_0, 10^{-6} (MPa×dpa)$^{-1}$**	**σ_θ, MPa measured (nominal)**	**Δd/d, %**	**ε^{ic}., %**	**B_0, 10^{-6} (MPa×dpa)$^{-1}$**	**σ_θ, MPa measured (nominal)**	**Δd/d, %**	**ε^{ic}., %**	**B_0, 10^{-6} (MPa×dpa)$^{-1}$**
0	0.1	0	0	0	0	0	0	0	0	0	0
59 (60)	0.3	0.2	0.74	49.8(50)	0.03	0.03	0.1	50.6(50)	0.19	0.19	0.56
59 (60)	0.3	0.2	0.74	50.1(50)	0.08	0.08	0.26	52.3(50)	0.25	0.25	0.72
0 (120)	0.1	0	0	94.6(100)	0.19	0.19	0.33	97.4(100)	0.45	0.45	0.69
69.5 (120)	0.6	0.5	1.57	94.4(100)	0.17	0.17	0.3	97.4(100)	0.40	0.40	0.61
130 (230)	1.1	1.0	1.68	190.8(200)	0.55	0.55	0.47	196.5(200)	0.97	0.97	0.74
141 (230)	1.2	1.1	1.7	190.5(200)	0.59	0.59	0.51	196.5(200)	0.97	0.97	0.74

TABLE 3 - *Irradiation creep characteristics of EP-852 F/M steel*

305°C/69 dpa					310°C/61 dpa				
σ_θ, MPa (nominal)	**σ_θ, MPa (measur.)**	**Δd/d, %**	**ε^{ic}, %**	**B_0, 10^{-6} (MPa×dpa)$^{-1}$**	**σ_θ, MPa (nominal)**	**σ_θ, MPa (measur.)**	**Δd/d, %**	**ε^{ic}, %**	B_0, 10^{-6} (MPa×dpa)$^{-1}$
0	0	0	0	0	0	0	0.1	0	0
50	38.5	0.05	0.05	0.25	60	56.0	0.15	0.05	0.19
50	38.2	0.03	0.03	0.15	60	58.0	0,3.	0.2	0.75
100	100.5	0.1	0.1	0.19	120	0	0.35	0.25	-
100	99.5	0.1	0.1	0.19	120	77.0	0.45	0.35	0.99
200	143.8	0.25	0.25	0.33	230	0	0.45	0.35	-
200	143.0	0.25	0.25	0.33	230	35.0	0.35	0.25	1.56

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